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Research Article

A Study on Modern High-Effective Random Packings for Ethanol-Water Rectification

Raschig Super-Ring is a modern and high-efficient packing used for intensification of absorption and distillation processes. The aim of this work is to characterize the efficiency of this packing applied to rectification of an important industrial system, ethanol-water, and to compare its efficiency to that of some random packings of the third generation as well as to the structured packing, HOLPACK, which is used in the ethanol production industry. The experiments were carried out in a column installation, 0.213 m in diameter with a packing height of 2.8 m. The column is heated by a number of electrical heaters (total power 45 kW), which can be switched gradually. Operation at total and partial reflux is possible. Eight types of random packings were studied: five types of Raschig Super-Ring, four metallic (with characteristic dimensions 0.5, 0.6, 0.7, and 1") and one of plastic material 0.6"; two types of packing IMTP and one plastic Ralu Flow. Some experiments were conducted at total reflux operation at vapor velocity, 0.253–0.936 m/s, and liquid superficial velocity, $4.44 \cdot 10^{-4}$–$1.63 \cdot 10^{-3}$ $m^3/(m^2 s)$. Experiments at partial reflux were carried out at constant liquid superficial velocity and changeable vapor velocity as well as at constant vapor velocity and changeable liquid velocity. The results are presented as height of transfer unit, HTU, and height equivalent to a theoretical plate, HETP, as a function of the velocity of phases.

Keywords: Distillation, Mass transfer, Packed bed columns, Raschig rings

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1 Introduction

The development of random packings has resulted in design of third generation packings created at the end of the 1970s characterized by small pressure drops, high efficiency, and high-technology design, for example Nutter Ring, CMR Ring, IMTP Ring etc. [1]. They surpass the performance of traditional random packings of second generation, Pall Ring and Intalox Saddle, immensely. Very popular is IMTP (Intalox Metal Tower Packing), used mainly for distillation processes but also very good for absorption, liquid-liquid extraction, and direct heat exchange [2]. Even better is Raschig Super-Ring [3], introduced in 1995, which is considered as a random packing of fourth generation. This packing is extensively used in the petrochemical industry for vacuum rectification processes, absorption, extraction, etc.[1]. For this reason, it is the subject of studies for pressure drop, effective surface area at large intervals of liquid throughput [4], and mass transfer controlled by gas or liquid phase [1, 5]. Concerning rectification, only a limited number of model systems is studied, for example, cyclohexane-*n*-heptane and iso-butane-*n*-butane [1].

The industrial importance of the ethanol-water system has recently grown in connection with the production of bio-ethanol for fuel. It is of interest to test the efficiency of modern random packings for rectification of this system.

The purpose of this work is to study the rectification of the ethanol-water system with three types of modern random packings – IMTP, Raschig Super-Ring, and Ralu Flow [6] at conditions close to real industrial operation. The experiments are carried out at high and medium concentrations. Most part of the column packing operates in this concentration range because of phase equilibrium peculiarity. In order of comparison, the structured metal packing, HOLPACK [7], largely used in ethanol-water rectification installations, is also studied at the same conditions. The investigated packings are presented in Fig. 1.

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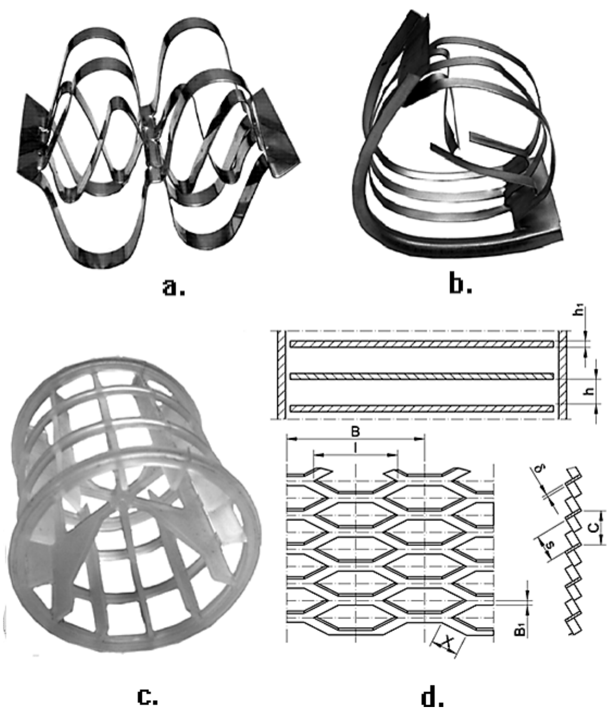

Figure 1. Photographs of the packings investigated.

2 Experimental Installation and Method of Study

The scheme of the experimental installation is shown in Fig. 2. It consists of a column (3), condenser (4), connecting pipelines, devices for monitoring (13) and measurement, and control panel (14). The column body is made of stainless steel with a bottom reboiler (1) with a volume of about 80 L. Electric resistance heaters (2) are incorporated into the reboiler with a total power of 45 kW. The inner diameter of the column body is 0.213 m. It is built of separate sections connected by flanges. The packing layer (3) of height 2.8 m is placed on a supporting grid. The elements of HOLPACK packing, made of expanded metal sheets, are arranged at constant distance using vertical distance elements of fixed height. In order to limit the harmful effect of irregular flow distribution towards the column wall and for amelioration of radial liquid distribution, reflecting rings (11) are mounted at a 200 mm-distance along the packing height [8]. They are also used in random packing experiments. A shower-type liquid distributor (10) is placed at the upper part of the column. It has 21 holes of 3 mm with Teflon nozzles of 1.7 mm. In order to avoid clogging, a filter is put before the distributor.

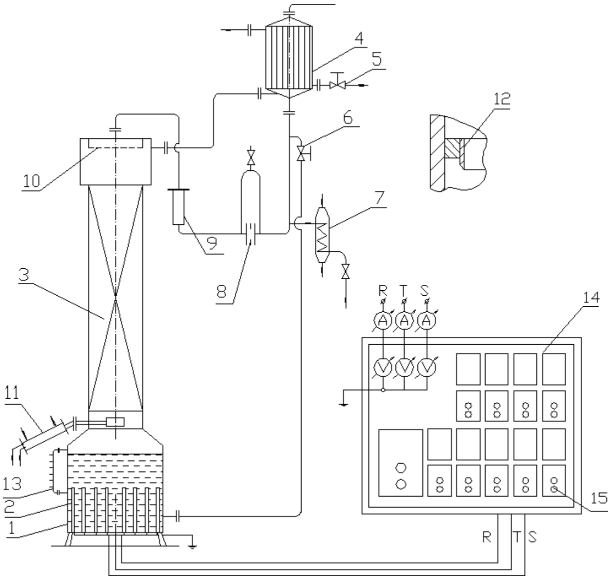

Figure 2. Scheme of the experimental installation.

The reflux condenser with water as cooling agent is a tubular heat exchanger made of stainless steel. The vapors pass and condense in the tubes. A diaphragm (8) and differential manometer are used for measuring reflux flow. The reflux can be partially or fully returned to the bottom, thus operating at total or partial reflux regime.

The column is thermally insulated with a 50 mm-layer of glass fibers. An equation has been obtained for determination of power losses, ΔP[1], based on the feeding power and the quantity of reflux flow at total reflux:

$$\Delta P = 2.081 + 0.0487\,P \tag{1}$$

The experiments were carried out in the following way: About 60 L of the ethanol-water mixture is fed into the column reboiler. The cooling water is turned on and the electric heating is switched on at full power. After the liquid is boiled, the electric heating is turned to minimal power and the warming of the whole column is awaited. The minimal flowrate of the liquid distributor necessary to ensure good distribution of liquid flow in the column was found experimentally (58 L/h). At this base, the minimal power of the heater was determined (13 kW). Because of the three-phase electric installation, at least three electric heaters have to be switched on or off simultaneously. So, the heating power can be changed in a stagewise manner. After each change, about 10 min are necessary for process stabilization. Samples are then taken before and after the packing. The electric power for every operational regime is derived from measurements of the current tension and strength. All runs were repeated twice, first increasing the power from minimal to maximal, then reducing the power from maximum to minimum.

The installation design allows for operation at variable reflux. It is possible to maintain constant liquid superficial velocity and change the vapor flow or maintain constant vapor flow and change the liquid flow, which is realized by returning the reflux flow in the bottom partially.

An alcohol densitometer with accuracy 0.1 % v/v was used for determination of sample concentration applying temperature correction, too. The mass concentrations were taken from

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[1] List of symbols at the end of the paper.

standard tables at 20 °C and the molar concentration was determined by the expression:

$$x = \frac{x_M}{2,56 - 1,56\,x_M} \tag{2}$$

Eight types of random packings have been studied: five types of Raschig Super-Ring made of metal (RSRM), one Raschig Super-Ring made of plastic (RSRP); two types of packing IMTP, and one plastic Ralu Flow, as well as the structured packing HOLPACK. The main parameters of the studied random packings are given in Tab. 1 and the geometrical characteristics of HOLPACK – in Tab. 2.

Each experiment with a packing was repeated at least twice or more if a considerable shift in the registered data has been observed. It has been shown experimentally with one type of packing that its redumping has no effect on the results.

3 Experimental Results

The results are presented as the mean overall mass transfer coefficient, $K_G\,a$, expressed by the driving force in vapor phase, Height of a Transfer Unit, HTU, and Height Equivalent to a Theoretical Plate, HETP, as a function of the gas capacity factor, F_V:

$$F_V = w\sqrt{\rho_G} \tag{3}$$

$K_G a$ is determined by integrating the mass transfer equation for the plug flow model:

$$w\,dy = K_G\,a\,(y^* - y)\,dh \tag{4}$$

$$K_G\,a = \frac{w}{H}\int_{y_W}^{y_D} \frac{dy}{y^* - y} \tag{5}$$

According to the method of transfer units, the apparatus height, H, is determined by the relation:

$$H = \mathrm{HTU}\ \mathrm{NTU} \tag{6}$$

Table 1. Main characteristics of the random packings investigated.

Type of packing	Material	Nominal diameter, [mm]	Surface area, [m^2/m^3]	Free volume, [%]
RSRM 0.5	metal	20	250	97
RSRM 0.6	metal	23	200	98
RSRM 0.7	metal	25	180	98
RSRM 1	metal	30	150	98
RSRP 0.6	plastic	23	200	93
IMTP 25	metal	25	200	96
IMTP 40	metal	40	150	97
Ralu Flow 1	plastic	25	165	95

Table 2. Geometric characteristics of HOLPACK packing in mm.

l	B	X	C	B_1	s	d	h_1	h
19	27	6.7	7.7	2.1	5.3	0.6	3.5	20

where NTU is the number of transfer units:

$$NTU = \int_{y_W}^{y_D} \frac{dy}{y^* - y} \tag{7}$$

and HTU can be obtained from the expression:

$$HTU = \frac{H}{\mathrm{NTU}} = \frac{w}{K_G\,a} \tag{8}$$

The values of NTU (Eq. (7)) are computed by numerical integration.

The results are also presented in terms of HETP, which represents the ratio of packing height and the number of theoretical plates. It is very convenient for efficiency comparisons of different packings.

The equilibrium concentration is calculated by the well-known equation:

$$y^* = \frac{\alpha x}{(\alpha - 1)\,x + 1} \tag{10}$$

with coefficient of relative volatility, α:

$$\alpha = \frac{P_A\,\gamma_A}{P_B\,\gamma_B} \tag{11}$$

The pure component vapor pressure is determined by the Antoine Equation [9]:

$$\log P = A - \frac{B}{C + t} \tag{12}$$

The constants A, B, and C are taken from [10] for vapor pressure at three temperatures – 70, 90, 100 °C, and are presented in Tab. 3.

The activity coefficients are determined by the relations [10]:

$$\ln \gamma_A = 1.58\,(1 - x)^3 \tag{13}$$

$$\ln \gamma_B = 0.79\,(3 - 2\,x)\,x^2 \tag{14}$$

The equations used for equilibrium calculations in the ethanol-water system correspond to experiments with an error lower than 0.5 % [10].

3.1 Experimental Results for Total Reflux Regime

The results at total reflux are shown in Figs. 3–7. An evident result is that packings with larger specific surface area are more efficient (Figs. 3–4). The highest values of mass transfer coefficient, K_G

Table 3. Antoine constants for the ethanol-water system.

Component	A	B	C
Ethanol	10.021	1459.1	212.69
Water	10.156	1705.9	231.22

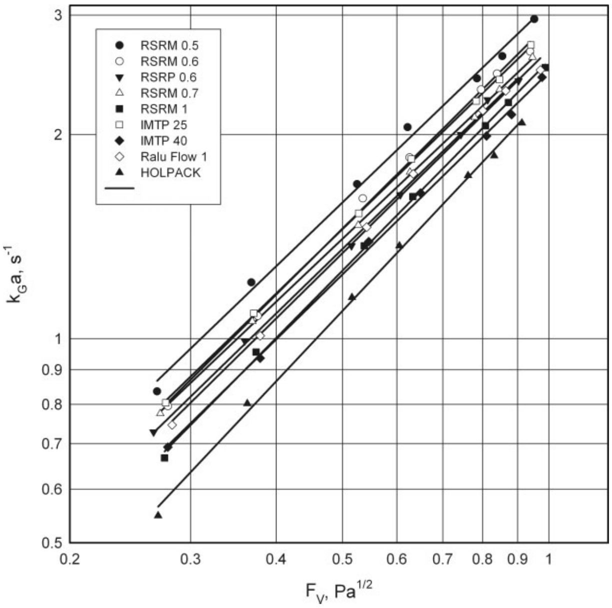

Figure 3. Mass transfer coefficient vs. F-factor for all packings.

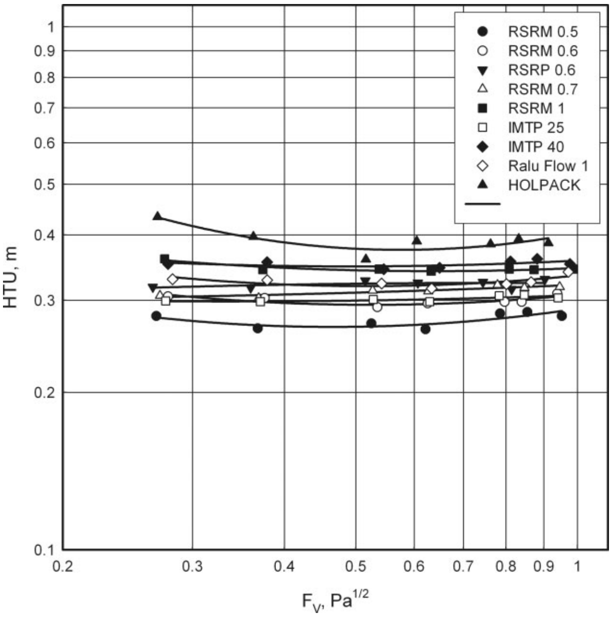

Figure 4. HTU vs. F-factor.

a, the smallest values of HTU and HETP, respectively, are obtained with RSRM 0.5 with a specific surface area 250 m^2/m^3, while the lowest coefficient, K_G *a*, is shown by HOLPACK packing with specific surface area 56 m^2/m^3. Regarding random packings, the lowest is the efficiency of RSRM 1 and IMTP 40 (specific surface area 150 m^2/m^3). There is no significant difference between the efficiency of HOLPACK packing and random packings although their specific surface areas are quite different. The reason is that in the case of HOLPACK packing, a great part of the mass transfer takes place in the space between sheets. In order to make a correct comparison, it is necessary to compare packings with approximately equal specific surface area.

Let us consider RSRM 1 and IMTP 40 packings (Fig. 5). The study is conducted in the main zone of operation before the loading point, where HETP is nearly constant and its mean value can be determined. It is equal to 0.347 m for IMTP 40. The packing RSRM 1 with its HETP of 0.339 m is 2.3 % more efficient.

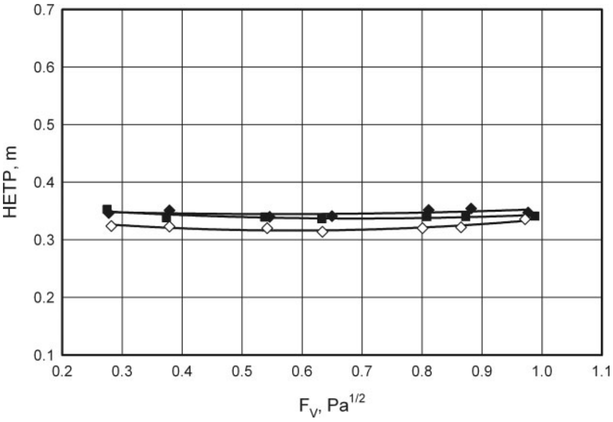

Figure 5. HETP vs. F-factor for RSRM 1, IMTP 40, Ralu Flow 1.

Other packings with equal specific surface area are RSRM 0.6 and IMTP 25. They do not show significant difference of HETP (Fig. 6).

It is interesting to compare RSRM 0,6 and RSRP 0,6 because they have the same dimensions but are made of different materials, metal and plastic. It is known that plastic packings are

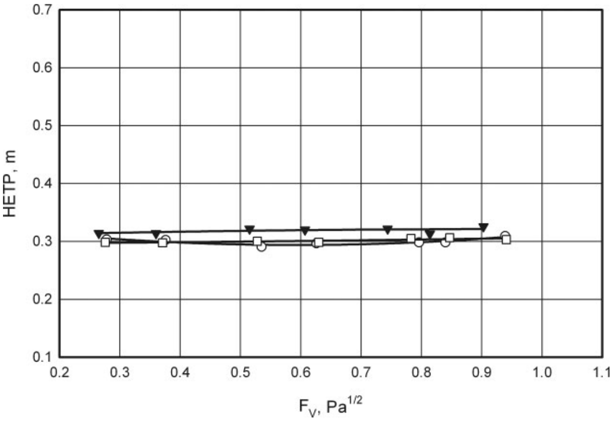

Figure 6. HETP vs. F-factor for RSRM 0.6, IMTP 25, RSRP 0.6.

less efficient due to their lower wetting. However, the difference between the metal and plastic RSR packing is found to be as low as 5.9 % (Fig. 6). This result shows that the Raschig company technology for improved wettability of plastic packings by covering with an additional layer [6] is rather successful.

Ralu Flow 1 has a specific surface area of 165 m^2/m^3, which is 10 % larger than that of RSRM 1 and IMTP 40. Its efficiency is 4.9 % and 7.1 % greater than the efficiency of the above packings, respectively (Fig. 5).

Fig. 7 illustrates the dependence of HETP on the F-factor for the rest of tested packings. The mean value of HETP for RSRM 0.5 is 0.278 m, which is a very good value for a random packing. For HOLPACK, this value is 0.388 m.

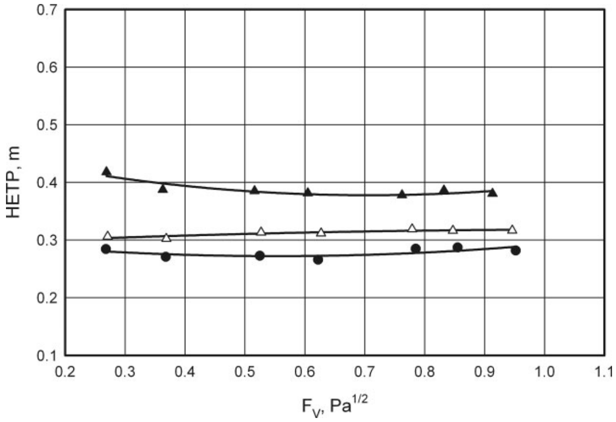

Figure 7. HETP vs. F-factor for RSRM 0.5, RSRM 0.7, HOLPACK.

3.2 Experimental Results for Regimes with Partial Reflux

Partial reflux is realized in two ways: maintaining constant vapor load and variable liquid load or maintaining constant liquid load and variable vapor load.

Fig. 8 illustrates the regimes with constant vapor velocity and variable liquid superficial velocity. The efficiency expressed as HTU is shown as depending on the ratio of *L/G*. It is seen that when reducing the ratio *L/G*, the efficiency initially rises then abruptly goes down.

For the case of constant liquid superficial velocity (Fig. 9), the efficiency increase is not so pronounced. However, as in the previous case, there is a steep efficiency reduction at lower *L/G* values. This effect is also confirmed by other authors [11].

Figs. 10–11 present the same results expressed as HETP depending on *L/G* ratio. The general curve trend is the same although less pronounced than in case of HTU-*L/G* dependence.

The results for HOLPACK packing are listed in more details because of the existence of mathematical models for the determination of its efficiency expressed as an overall mass transfer coefficient and HTU. For regimes with constant vapor and variable liquid load, the experimental points are systematically shifted from the model prediction (Fig. 12). It might be attributed to the large rate of axial mixing (50–60 %), which is not

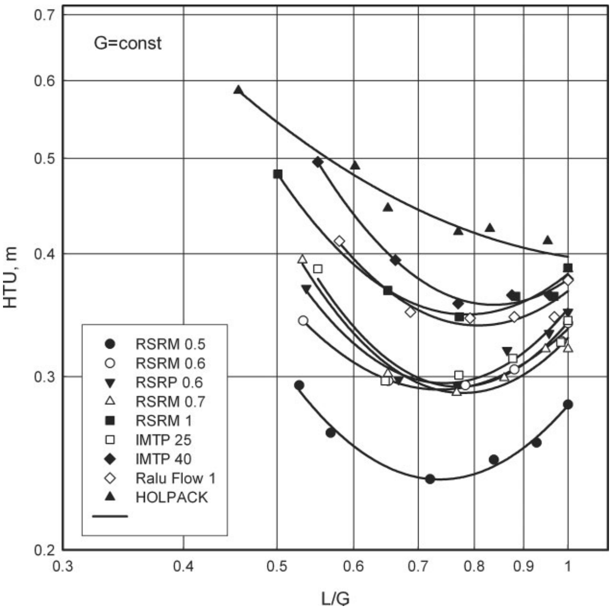

Figure 8. HTU vs. the ratio *L/G* at constant vapor load.

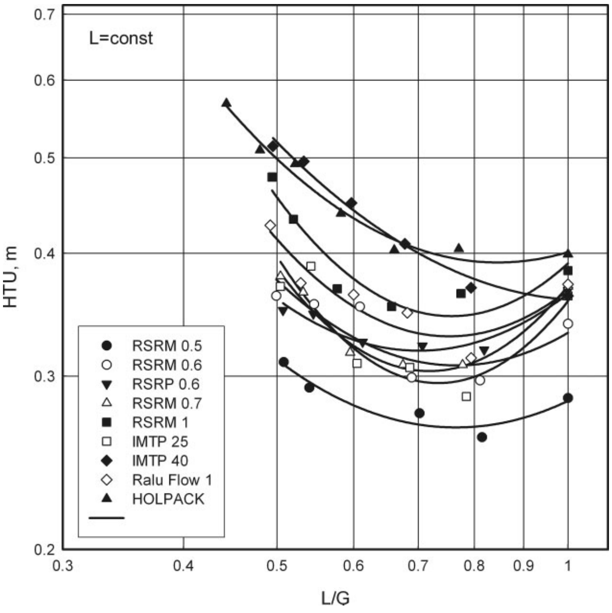

Figure 9. HTU vs. the ratio *L/G* at constant liquid load.

precisely accounted for by the model. At lower *L/G* values, the shift becomes larger.

Regarding regimes with constant liquid and variable vapor throughput (Fig. 13), the experimental and model results coincide for *L/G* values in the zone 0.5–1. An explanation might be based on the fact that the liquid superficial velocity is low and the influence of the axial mixing, being manifested only in liquid phase, is not changed significantly. However at *L/G* values lower than 0.5, the experimental values of HTU again lie over the model prediction.

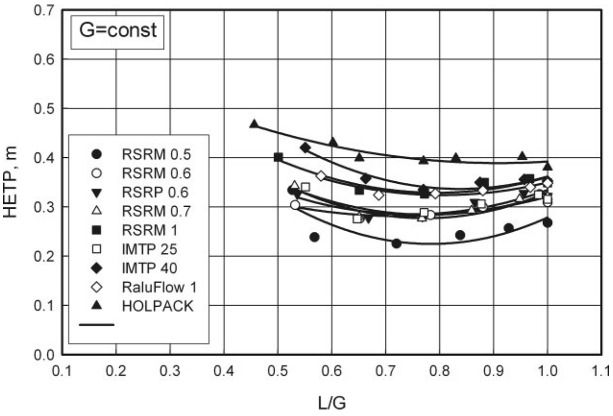

Figure 10. HETP vs. the ratio *L/G* at constant vapor load.

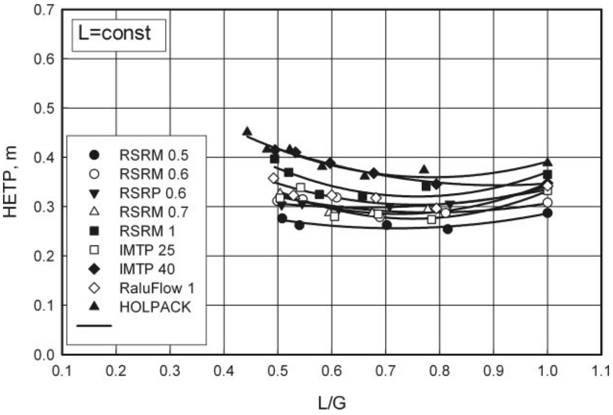

Figure 11. HETP vs. the ratio *L/G* at constant liquid load.

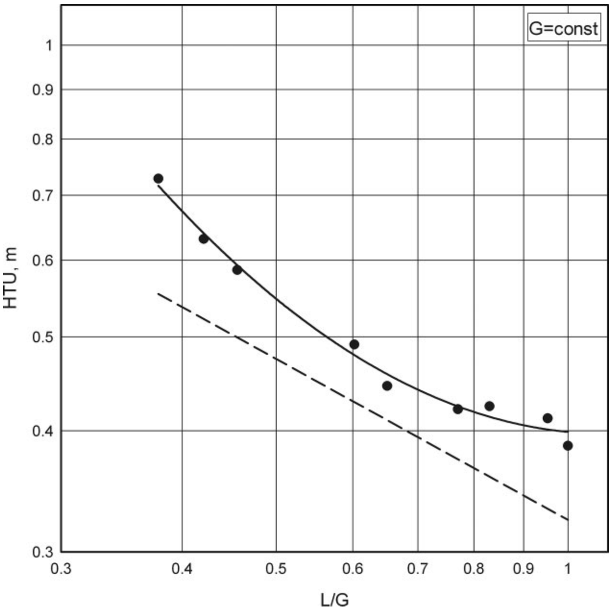

Figure 12. HTU vs. the ratio *L/G* at constant vapor load, HOLPACK.

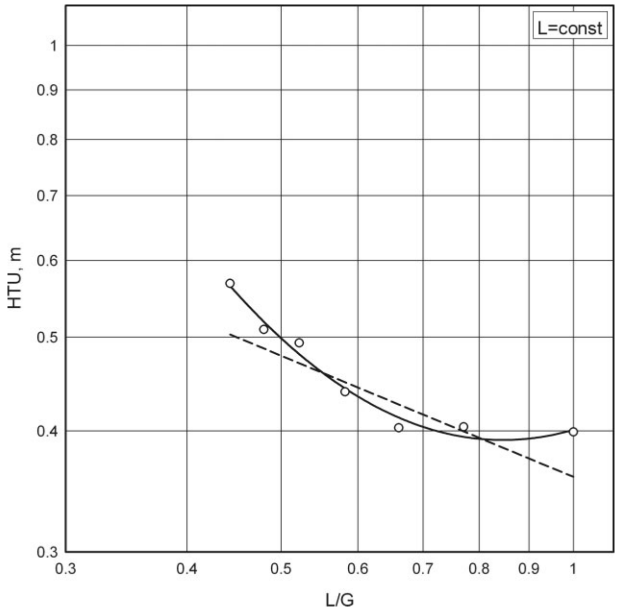

Figure 13. HTU vs. the ratio *L/G* at constant liquid load, HOLPACK.

As far as the model determining the volume mass transfer coefficient accounting only for some phenomena with major impact on the mass transfer (axial mixing and Marangoni effect), it should be supposed that other phenomena influence the efficiency at low *L/G* ratio. In our opinion, the efficiency reduction in this case is due to the irregular flow distribution, which causes changes of the operating line resulting of crossing in some points with the equilibrium line. The influence of irregular flow distribution is the subject of a future study.

4 Conclusion

The results of this study on packing performance in ethanol-water rectification demonstrate very good efficiency of random packings. The best is the smallest dimension of Raschig Super-Ring with a mean value of HETP = 0.28 m. It is 28 % better than the structured packing, HOLPACK, used currently in the ethanol production industry.

Comparing the metal and plastic packings with equal specific surface area (RSRM 0,6 and RSRP 0.6), up to 6 % lower efficiency of the plastic packing is found. These results certify that a shortcoming of plastic packings (low wetting) is rather overcome.

The results of runs at variable reflux show an initial increase of efficiency with the reduction of liquid/vapor flow ratio until a certain value and abrupt efficiency reduction thereafter.

Symbols used

F_V	$[Pa^{1/2}]$	F-factor
G	$[mol/(m^2s)]$	vapor molar flow

H	[m]	column height
HETP	[m]	Height Equivalent to a Theoretical Plate
HTU	[m]	Height of a Transfer Unit
$K_G\,a$	[1/s]	overall mass transfer coefficient
L	[mol/(m^{2}s)]	liquid molar flow
NTU	[–]	Number of Transfer Units;
P	[kW]	feeding power in Eq. (1)
P_A	[Pa]	saturated vapor pressure of pure ethanol
P_B	[Pa]	saturated vapor pressure of pure water
t	[°C]	temperature
w	[m/s]	mean vapor velocity based on column cross-section
x	[mol/mol]	ethanol molar concentration in liquid phase
x_M	[kg/kg]	ethanol mass concentration in liquid phase
y	[mol/mol]	ethanol molar concentration of vapors
y^*	[mol/mol]	equilibrium vapor concentration
y_W	[mol/mol]	initial vapor concentration
y_D	[mol/mol]	final vapor concentration
a	[–]	coefficient of relative volatility
γ_A	[–]	activity coefficients of ethanol
γ_B	[–]	activity coefficients of water
ρ_G	[kg/m^3]	vapor density

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